



Practitioner's Docket No. DP-304112 (DEP-0243)

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RECEIVED  
JUL 14 2003  
GROUP 1700

APPLICANT: ISMAT ABU-ISA )  
SERIAL NO.: 10/055,112 ) Examiner:  
FILED: JANUARY 23, 2002 )  
FOR: ELASTOMERIC INTEMUSCENT ) Art Unit:  
MATERIAL )

Commissioner for Patents  
Alexandria, VA 22313-1450

**PETITION UNDER 37 CFR 1.59(b) TO EXPUNGE ACCOMPANYING INFORMATION  
SUBMITTED UNDER M.P.E.P. § 724.02**

SIR:

Applicant hereby petitions under 37 CFR § 1.59(b) to expunge the information submitted herewith under MPEP § 724.02. The information which is submitted herewith and for which Applicant petitions to be expunged is proprietary, and the information has not been otherwise made public to the knowledge of the Applicant. Petitioner confirms that the Applicant will retain the information submitted herewith for the period of any patent with regard to which this information is submitted. This petition is being submitted by the party in interest who originally submitted the information. The fee required by 37 CFR § 1.17(h) is enclosed. If additional charges are incurred with respect to this Petition or otherwise, they may be charged to Deposit Account 06-1130 maintained by Cantor Colburn LLP.

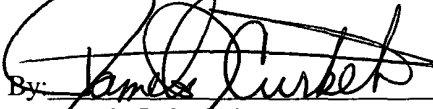
07/10/2003 GWORDOF1 00000115 10055112

01 FC:1460

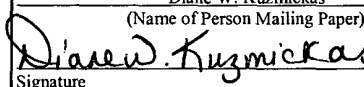
130.00 DP

Respectfully submitted,

CANTOR COLBURN LLP

By:   
Pamela J. Curbelo  
Registration No. 34,676

Date: July 7, 2003  
Address: 55 Griffin Road South, Bloomfield, CT 06002  
Telephone: (860) 286-2929

I hereby certify that this correspondence was deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on	
July 7, 2003 (Date of Deposit)	
Diane W. Kuzmickas (Name of Person Mailing Paper)	
	07/07/03 Date

\$1714



Practitioner's Docket No. DP-304112 (DEP-0243)

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT: ISMAT ABU-ISA )  
SERIAL NO.: 10/055,112 ) Examiner:  
FILED: JANUARY 23, 2002 )  
FOR: ELASTOMERIC INTEMUSCENT ) Art Unit:  
MATERIAL )

RECEIVED  
JUL 14 2003  
GROUP 1700

Commissioner for Patents  
Alexandria, VA 22313-1450

**TRANSMITTAL OF PROPRIETARY MATERIAL NOT OPEN TO THE PUBLIC, TO BE  
OPENED BY EXAMINER OR OTHER AUTHORIZED PTO EMPLOYEE  
(M.P.E.P. § 724.02)**

SIR:

Applicant hereby submits the attached material reference A, which is considered to be proprietary and Applicant requests that this material be considered under M.P.E.P. § 724.02. A Petition under 37 CFR § 1.59 and the fee therefore (37 CFR § 1.17(h)) to expunge the information, if found not to be important to a reasonable Examiner in deciding whether to allow the Application to issue as a patent, also accompanies this material. If additional charges are incurred with respect to this Transmittal or otherwise, they may be charged to Deposit Account 06-1130 maintained by Cantor Colburn LLP.

Respectfully submitted,

CANTOR COLBURN LLP

By: 

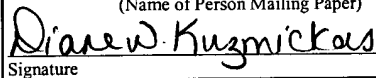
Pamela J. Curbelo  
Registration No. 34,676

Date: July 7, 2003  
Address: 55 Griffin Road South, Bloomfield, CT 06002  
Telephone: (860) 286-2929

I hereby certify that this correspondence was deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on

July 7, 2003  
(Date of Deposit)

Diane W. Kuzmickas  
(Name of Person Mailing Paper)

 07/07/2003  
Signature Date

**CERTIFICATE OF MAILING BY FIRST CLASS MAIL (37 CFR 1.8)**

Applicant(s): Ismat Abu-Isa

Docket No

DP-304112 (DEP-0243)

Serial No.

10/055,112

Filing Date

January 23, 2002

Examiner

Group Art Unit

Invention: ELASTOMERIC INTEMUSCENT MATERIAL

**RECEIVED**  
JUL 14 2003  
GROUP 1700

Transmittal of Proprietary Material to be Opened by Examiner or Other Authorized PTO Employee (1 pg.); Petition to Expunge Accompanying Information (1 pg.); Proprietary Information (Reference A)(29 pgs.); Certificate of Mailing by First Class Mail (1 pg.); Check for Fees

I hereby certify that this

*(Identify type of correspondence)*

is being deposited with the United States Postal Service as first class mail in an envelope addressed to

Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on July 7, 2003*(Date)*Diane W. Kuzmickas*(Typed or Printed Name of Person Mailing Correspondence)*  
*(Signature of Person Mailing Correspondence)*

Note: Each paper must have its own certificate of mailing.

P07A/REV03

DP-302280

RECEIVED  
JUL 14 2003  
GROUP 1700INTUMESCENT MATERIAL USING HIGH DENSITY POLYETHYLENE  
REGRIND

## TECHNICAL FIELD

This invention relates to fire protection materials, and, more particularly, to a moldable intumescent composite material made from recycled plastic.

5

## BACKGROUND OF THE INVENTION

Various methods exist in the prior art for the protection of persons and property against fire. Among these methods is the use of intumescent materials, which are generally fire retardant composite compounds that swell and form ceramic-like sponges when exposed to the high temperatures generally associated with fires. Intumescent materials are typically incorporated into coatings or paints and are most often formed into mat-like structures that may be placed between a high fire risk apparatus and an apparatus or area for which fire protection is desired.

10

Intumescent coatings are generally used on the wall surfaces of buildings, ships, aircraft, motor vehicles, or other vessels intended for human occupancy and which are susceptible to fires. The nature of the coating allows its thickness to be kept at a minimum until it is activated by high temperatures. Such coatings contain film-forming resins that, upon being subjected to high temperatures, undergo molecular changes that result in corresponding changes in physical properties such as a reduced ability to conduct heat. The coatings also contain ingredients that will react with each other upon heating to generate gases and form an incombustible or low combustible residue. The expelled gases expand the residue into a foam having thermal insulating properties. In many instances a major part of the residue formed is a carbon material that is formed by the dehydration of a polyhydric substance such as a polyalcohol. The

20

25

DP-302280

reactions of the ingredients occur within the coating so that when the coating film is heated to beyond a specific elevated temperature, intumescent additives (such as the polyhydric alcohol, ammonium hydrogen phosphate or hydrated magnesia) are decomposed, thereby generating water, carbon dioxide, ammonia, and/or other heat absorbing gases that do not readily support combustion. However, although intumescent coatings are readily available, they require a lengthy and complicated process for their application to a surface. Oftentimes, a primer is required to promote adhesion to the surface of the structure the coatings are intended to protect. A drying time, sometimes as long as many hours, is required for the primer and the coating. When these coatings are exposed to fire and intumesce, they are transformed into a weak sponge that cannot withstand raging drafts and thermal expansion forces that occur as a result of a fire, and, therefore, they oftentimes provide only limited protection for a short period of time. A distinct advantage of the intumescent material described in this application over intumescent coatings is that the intumescent material can be formed into stand alone parts.

Intumescent mat-like structures are often used in place of coatings to avoid the problems associated with the coatings. In the mat-like structures, multiple layers of fire resistant materials having intumescent materials located therebetween are used to provide a flexible wrap that provides stability to expanded intumescent materials. These flexible wraps generally include alternate layers of fire resistant materials and intumescent materials that are designed to expand to provide a consistent and effective fire protection system. Although mat-like structures can be manufactured to have sufficient flexibility that allows them to conform to the shape of the object to be protected (e.g., conduits, electrical cable, etc.), they tend to require additional hardware or an adhesive in order to be held into place.

Because of the above-mentioned drawbacks associated with intumescent coatings and mats generated from these coatings, it would be

DP-302280

desirable to provide a moldable thermoplastic composition having intumescent properties. Such moldable thermoplastic compositions could be shaped to adapt to the particular application at hand, and may also serve as an integral part of the structure being protected. For example, the intumescent material can be easily  
5 introduced as the outermost layer of a multi-layer fuel tank to provide fire-shielding protection for the tanks. This is done as the layers are being formed together in an extrusion process to form a multi-layer sheet or parison used for thermal forming or blow molding of the tank. No added steps are required to incorporate the intumescent material into the structure. Similarly, intumescent  
10 material can be extruded over a cable to provide electrical insulation and fire protection simultaneously.

#### SUMMARY OF THE INVENTION

An intumescent composite material typically contains ground  
15 recycled plastic, chlorinated polyethylene, a plasticizer, foaming agents, a char formation compound, an antioxidant, hydrated oxides, smoke-reducing compounds, and filler material. A cross-linking agent may also be incorporated into the material to improve the rigidity of the material when it is exposed to fire.

The invention also includes a method of improving the  
20 processability of the intumescent composite material during its formulation. The method comprises adding titanium dioxide and silicone rubber to the material and molding the material to a desired shape.

#### BRIEF DESCRIPTION OF THE DRAWINGS

25 The apparatus and method will now be described by way of example, with reference to the accompanying Figures, which are meant to be exemplary and not limiting.

DP-302280

Figure 1 is a perspective view of an intumescent flammability test apparatus used for measuring the efficiency of moldable intumescent composite materials of the present invention.

Figure 2 is a graph depicting temperature versus time at different locations of a steel plate protected by the moldable intumescent composite material of the present invention.

#### DETAILED DESCRIPTION OF THE INVENTION

This invention relates to an intumescent thermoplastic molding composition that has as its primary constituent recycled plastic. In one embodiment, the recycled plastic obtained is ground from unwanted plastic articles, such as scrap generated during the manufacturing of plastic fuel tanks or from regrind obtained from post consumer plastic fuel tanks, milk bottles, garbage bags, or other plastic containers made of high density polyethylene. The ground plastic (hereinafter referred to as "regrind") can be molded in a variety of ways using thermal processes to produce useful products. The resultant product is an intumescent composite material that can be molded to serve as a heat or fire barrier. The particular geometry or configuration of such a product is dependent upon the particular application of the material. Oftentimes an article molded from the intumescent composite material serves only incidentally as a heat or fire barrier and has an altogether different functional purpose. For example, the material may be used by itself or as a part of a composite structure to mold vessels for the containment of fluids or solids, or it may be used to form panels, dashboards, consoles, or other parts for use in automobile, aircraft, or marine applications. Other uses will be apparent to those skilled in the art. Whether used for heat or fire protection or not, the material provides physical and fire-resistant properties that are comparable to those of similar compositions utilizing virgin high density polyethylene (HDPE) as the base material.

DP-302280

A preferred embodiment of the intumescent thermoplastic molding composition includes regrind (e.g., recycled HDPE or the like) with chlorinated polyethylene (CPE) and/or silicone rubber, an intumescence-inducing foaming agent, and a char formation compound. The composition may also include

5 antioxidants, a plasticizer, smoke reducing compounds, and fillers. The formulation of the intumescent composite material on a laboratory scale can be achieved in a variety of ways including, but not being limited to, mixing using a two-roll mill, a Brabender bowl, a Banbury mixer, a Brabender extruder, a twin-screw extruder, or the like, all of which are well known in the art. The

10 formulation of the material on a plant scale is typically achieved using a single-screw kneader or similar melt mixing devices designed to produce low shear during mixing.

Referring now to Table 1, suitable ranges for the formulation of the intumescent composite material are illustrated. The "composition" columns

15 encompass a variety of suitable compositions in which the HDPE and the CPE can be used in combination with each other. These compositions are based upon the total weight of the intumescent material. In other embodiments, it will be shown that a suitable silicone rubber may be substituted for some or all of the CPE in a formulation.



DP-302280

TABLE 1 Intumescent Material Formulation			
INGREDIENT	Composition (wt %)	Preferred Composition (wt %)	More preferred Composition (wt %)
Regrind	about 2 to about 60	about 5 to about 45	about 15 to about 25
First Material: Chlorinated polyethylene	up to about 60	about 5 to about 45	about 27 to about 33
Silicone rubber	up to about 60	about 1 to about 30	about 2 to about 5
Foaming Agent: Phosphate-based	up to about 20	about 6 to about 18	about 7 to about 16
Hydrated magnesia	up to about 20	About 2 to about 19	about 15 to about 17
Char forming Compound:	up to about 20	about 1 to about 15	about 3 to about 10
Char forming Compound (smoke reducer): Antimony oxide	up to about 8	about 0.5 to about 6	about 1 to about 5
Optional Ingredients:			
Plasticizer	up to about 15	about 3 to about 12	about 5 to about 10
Antioxidant: Sulfur-based	about 0.1 to about 5	about 0.2 to about 1	about 0.4 to about 0.6
Phenol-based	about 0.1 to about 5	about 0.2 to about 1	about 0.4 to about 0.6
Filler	up to about 20	about 2 to about 18	about 5 to about 10
Smoke reducer: Titanium dioxide	up to about 5	about 0.5 to about 4	about 1 to about 3
Cross-linking agent	up to about 2	Up to about 1	up to about 0.1

The base ingredient of the intumescent composite material is the recycled HDPE. Regrinding and reusing this plastic allows for a significant cost savings to be realized in the manufacture of the intumescent composite material.

- 5    Regrind, in one embodiment used herein comprises about 2 weight percent

DP-302280

(wt%) or greater contaminants, with about 5 wt% to about 40 wt% preferred, and about 5 wt% to about 15 wt% especially preferred. In contrast, virgin HDPE comprises greater than about 99% purity. These contaminants can comprise thermoplastic materials (other than HDPE), thermoset materials, elastomers, fillers, adhesives, and combinations comprising at least one of the foregoing contaminants.

In an alternative embodiment, regrind used herein comprises HDPE which has been processed at elevated temperatures, e.g., heat treated via a molding process such as injection molding, blow molding, and the like. For example, the regrind may have been processed at temperatures exceeding about 150°C, with temperatures of about 150°C to about 190°C typical. In contrast, the virgin HDPE has not been heat treated, i.e. exposed to temperatures exceeding about 150°C, e.g., in the formation of an article.

For example, in one embodiment, the source of the HDPE is unwanted plastic liners of fuel tanks, although other sources may be considered. The chemical composition of regrind from fuel tank liners differs slightly from that of virgin HDPE primarily because fuel tank liners contain a copolymer of ethylene vinyl alcohol, which is used in the original manufacture of the tank liner as a gasoline permeation barrier. This regrind also contains maleated polyethylene adhesive. Although the ethylene vinyl alcohol and the maleated polyethylene are contaminants in the formulation that negatively affect the physical properties of the material, the composition containing regrind retains its properties. This is believed to be attributable to the incorporation of the CPE into the formulation, which buffers these negative effects and causes articles molded from compositions using the regrind to be of a quality that is comparable to that of articles molded from compositions using virgin HDPE. Because of the variation in the compositions of virgin HDPE and regrind, and because the vinyl alcohol and the polyethylene adhesive can be viewed as contaminants, the

DP-302280

substitution of regrind for virgin HDPE would not be obvious to a person of ordinary skill in the art.

5 The CPE, which typically has a chlorine content on a weight basis of about 25% to about 49%, with a content of about 32% to about 42% being preferred, and a content of about 36% to about 42% being most preferred, may be combined with regrind in any one of a number of different ratios to produce the intumescent material compound. The CPE for both formulations is used to tailor mechanical properties and improve flammability resistance, thus resulting in higher char formation after exposure to fire. The CPE serves as an impact  
10 modifier that will introduce rubber-like characteristics to the intumescent material, which serves to dampen or isolate noise and vibration. As stated above it also improves flammability characteristics and promotes char formation. Conventionally, CPE is used to make rubber goods for the automotive industry, such as hoses and seals, as well as for non-automotive applications (swimming  
15 pool liner, industrial hoses, wire coatings, and the like). The polymer is also used to make seals and o-rings because of its relatively low permeability to many fluids. The material is available as a commercial product, from the DuPont Dow Elastomers Company, Midland, Michigan.

The formulation also preferably incorporates various compounds  
20 in order to improve the low temperature properties and processability of the intumescent composite material. These various compounds typically include a plasticizer and/or silicone rubber. The plasticizer is typically a chlorinated wax compound in the form of a liquid. Other plasticizers include organic phosphates and organic esters. In a preferred embodiment, the plasticizer used is PAROIL  
25 145, which may be purchased from Dover Corporation, Industrial Division, Canada.

The foaming agent is incorporated into the intumescent composite material to foam the material after it is exposed to the high temperature of a fire. During a fire, high temperature causes the polymer matrix to soften, and at the

DP-302280

same time or shortly thereafter causes the evolution of gases from the foaming agent. The gases diffuse through the intumescent composite material as the material starts to soften. This diffusion forms air pockets, which make the material porous. The porosity of the material improves its thermal insulation characteristics. Foaming agents typically include either ammonium dihydrogen-  
5 ortho-phosphate ( $\text{NH}_4\text{H}_2\text{PO}_4$ ), an ammonium polyphosphate ( $(\text{NH}_4\text{PO}_3)_x$ ), as well as combinations comprising at least one of the foregoing. Also, because they emit water at the early stages of fire exposure, hydrated metal oxides and hydroxides, such as hydrated magnesium oxide, magnesium hydroxide, hydrated  
10 aluminum oxide, as well as combinations comprising at least one of the foregoing, are good foaming agents. In a preferred embodiment, the foaming agent of choice is PHOSCHECK P/30, an ammonium polyphosphate which may be obtained from Monsanto Company, St. Louis, Missouri.

Various other foaming agents can also be incorporated into the  
15 formulation. These foaming agents may be either hydrated alumina ( $\text{Al}_2\text{O}_3$ ), hydrated magnesia ( $\text{MgO}$ ), magnesium hydroxide ( $\text{Mg}(\text{OH})_2$ ), metal carbonates and bicarbonates (which emit carbon dioxide upon being heated), azides and other organic nitrogen compounds (which emit nitrogen upon decomposition), and the like, as well as combinations comprising at least one of the foregoing  
20 compounds. Hydrated alumina and hydrated magnesia emit their hydrated portions as water vapor, which has a tendency to reduce the temperature of the fire. Magnesia also has a tendency to absorb hydrochloric acid gas that is produced and evolved during the aging of the CPE, and further has the ability to act as a dehydrochlorination stabilizer. In a preferred embodiment, a  
25 combination of hydrated alumina and hydrated magnesia are used. The hydrated alumina is commercially available as MARTINAL OL-104C from Lonza, Inc., Fair Lawn, New Jersey, while magnesia is commercially available as MAGSHIELD 98 from Martin Marietta, Baltimore, Maryland, among other sources.

DP-302280

In order to protect the regrind/CPE from heat and oxygen, a suitable thermal and oxidation stabilizer is incorporated into the intumescent composite material. A system consisting of sulfur-based distearylthiodipropionate and a hindered phenol, which may be the butylated reaction product of p-cresol and dicyclopentadiene, is effective as an antioxidant and is the preferred thermal and oxidation stabilizer. Hindered phenols allow the addition of free radicals generated during polymer oxidation and deactivate them to slow down the oxidation process, while sulfur and amine compounds react in the termination step of the autocatalytic oxidation of polymers, thus changing active free radicals to inactive compounds, which slows down the oxidative degradation. The combination of both compounds has been found to produce a synergistic effect. Distearylthiodipropionate is commercially available as DSDTP from Witco Corporation, Greenwich, Connecticut, and the phenol is available as WINGSTAY L from R.T. Vanderbilt, Norwalk, Connecticut. Other antioxidants based on hindered phenols, such as Irganox 1010 from Ciba Geigy, or sulfur compounds, such as Vanax A from R.T. Vanderbilt, or amine containing compounds, such as Agerite White from R.T. Vanderbilt, are also effective thermal and oxidation stabilizers for the intumescent material.

A char formation material is also added to the intumescent composite material to form heavy char when the material is exposed to fire. Char is a black residue formed as a result of the burning of an organic material. Once formed, char becomes chemically stable and functions as a thermal barrier by inhibiting further combustion. The char formation material may be any suitable carbohydrate, polyhydric alcohols (such as pentaerythritols, and the like), as well as combinations comprising at least one of the foregoing materials. Starch or sugar derivatives are also effective char forming additives. A preferred char formation agent is a blend of monopentaerythritol and dipentaerythritol, which is commercially available as PERSTORP PE from Perstorp Compounds, Inc., Florence, Massachusetts.

DP-302280

The intumescent composite material can also comprise additives aimed at reducing smoke when the intumescent composite material burns. These additives are typically metal oxides such as antimony oxide, titanium dioxide, and the like, as well as combinations comprising at least one of the foregoing  
5 oxides. In addition to reducing smoke, these oxides also improve the processability of the intumescent composite material during formation by reducing the torque required for melt processing of the HDPE and the CPE. They have the further advantage of enhancing the aesthetic qualities of articles molded from the material by providing a cosmetically-superior surface finish on  
10 a microscopic level. These oxides also provide added flame retardancy to the formulation and serve to further enhance the formation of char. In a preferred embodiment, silicone rubber is also used to enhance char formation and reduce smoke. Other materials that may reduce smoke include cobalt and manganese organic compounds.

15 In a formulation in which both regrind and CPE are added, a silicone rubber may also be added to reduce the amount of smoke generated. Silicone rubber can be synthesized as a silphenylene siloxane elastomer or purchased as DOW SILICONE 4-7081 from Dow Corning, Midland, Michigan. In another alternate embodiment, silicone rubber can be substituted for CPE up  
20 to the full weight percent of CPE, thereby resulting in a finished product that will evolve a gaseous product upon burning that contains no chlorinated gas molecules, as shown in Example 2 below.

The intumescent composite material may contain cross-linking agents. Generally, these cross-linking agents are small concentrations of free  
25 radical-generating peroxides, such as dicumyl peroxide, and crosslinking co-agents, such as trimethylolpropane trimethacrylate (TMPT), and the like, as well as combinations comprising at least one of the foregoing cross-linking agents. The CPE used in the intumescent material formulation can also be cross-linked using sulfur or amine-based curatives. These agents improve the rigidity of the

DP-302280

intumescent composite material, especially upon exposure to fire. Dicumyl peroxide is commercially available as DICUP 40KE from Hercules Inc., Wilmington, Delaware, and TMPT is available as a liquid or adsorbed to the surface of silica powder (Saret 500) from Sartomer Company, Inc., Exton,  
5 Pennsylvania.

In appropriate formulations, a filler consisting of glass, mica, carbon (such as carbon black and the like), titanium oxide, clay, calcium carbonate, and the like, as well as combinations comprising at least one of the foregoing fillers, in the form of particles, fibers, powder, spheres, nanotubes,  
10 nanospheres, and the like, as well as combinations comprising at least one of the foregoing forms, can be added. These fillers help to strengthen the molding of the intumescent composite material during processing and allow the material to develop a strong structure after intumescenting. In a preferred embodiment, the filler is glass fiber, which is available as PPG CHOPPED GLASS from PPG  
15 Industrial Inc., Pittsburgh, Pennsylvania.

The main function of the moldable intumescent composite material formulated from the regrind is to resist the spread of flame from a fire source and to shield articles protected by the material from elevated temperatures. The characteristics of intumescence efficiency are measured by a  
20 procedure using an intumescent flammability test apparatus, shown generally at 10 in Figure 1. Moldable compositions illustrated in this specification have been tested as intumescent compositions to be used for resisting the spread of a flame from a fire source and curtailing temperature rise. The characteristics of intumescence efficiency as they relate to moldings made from the subject  
25 compositions were determined using intumescent flammability test apparatus 10 and by the practice as outlined below. When reference is made to such property in this specification or in the claims, it is intended that the property be determined utilizing intumescent flammability test apparatus 10 or an equivalent apparatus.

DP-302280

Intumescent flammability test apparatus 10 is a three-walled chamber comprising a left side wall 12, a back wall 14, and a right side wall 16. Each wall 12, 14, 16 is a steel plate 229 millimeters (mm) high, 127 mm wide, and 1 mm thick. A steel plate 152 mm square and 1 mm thick is used as a roof 18. The moldable intumescent composite material, shown generally at 20, is affixed to the under side of roof 18, while four thermocouples are welded on the top side of roof 18 at locations 22, 24, 26, 28 to monitor temperatures during the flammability test. Flame temperatures are measured using a thermocouple 30 inserted through side wall 12 and into the blue flame region of a Bunsen burner 32, which is used as the flame source. Bunsen burner 32 is 165 mm tall, and the flame height above the top of Bunsen burner 32 is 60 mm and is adjusted so that the tip of the inner blue cone of the flame touches the surface of intumescent composite material 20. Burning tests are carried out for 30 minutes.

Referring now to Figure 2, thermocouple temperature readings are plotted versus time of exposure to the flame. The flame temperature, shown at 130, correlates to the temperature read by thermocouple 30. In addition, the thermocouples at locations 22, 24, 26, 28 correlate to the temperatures plotted on the graph of Figure 2 at 122, 124, 126, 128, respectively. The temperatures are noted as a measure for the degree of effectiveness of intumescent composite material 20 in providing fire and heat shielding.

Referring now to Table 2, the formulations of various embodiments of intumescent composite compounds are illustrated as Samples 1 through 5. The formulations of Samples 3 and 4 incorporate virgin HDPE, and are shown for comparison only. The finished composite materials that are produced are moldable intumescent thermoplastic elastomers having varying degrees of hardness.



TABLE 2						
INGREDIENT	SAMPLE					
	Control C (wt. %)	Sample 1 (wt. %)	Sample 2 (wt. %)	Sample 3 (wt. %)	Sample 4 (wt. %)	Sample 5 (wt. %)
Virgin HDPE	23	-	-	23	23	-
Regrind	-	23	23	-	-	23
Chlorinated polyethylene	30	30	30	30	30	30
Silicone rubber	-	-	2	2	2	2
Chlorowax	7	7	7	7	7	7
Phosphate-based foaming agent	8	8	8	8	7	7
Hydrated magnesia	2	2	2	2	2	2
Hydrated alumina	15	15	15	15	15	15
Distearyl- Thiopropionate	0.5	0.5	0.5	0.5	0.5	0.5
Hindered Phenol	0.5	0.5	0.5	0.5	0.5	0.5
Filler	6	6	6	6	6	6
Antimony Oxide	3	3	5	5	5	5
Titanium dioxide	-	-	2	2	2	2
Pentaerythritol	5	5	5	5	5	5
Dicumyl peroxide		-	-	-	0.05	0.05
Trimethylolpropane Trimethacrylate		-	-	-	0.05	0.05
Torque* (m-g)	7208	5000	5117	3434	3302	4740

\*during melt mixing; meters-gram (m-g)

Referring now to Table 3, lower maximum temperatures measured across the thickness of intumescent composite material 20 as intumescent composite material 20 is exposed to the flame may imply improved shielding efficiency properties. Even though the flame of Bunsen burner 32 exceeds 1,000°C and the exposure time is 30 minutes, the maximum temperature

DP-302280

is only marginally higher (286°C) for formulation Sample 1, when compared to the 282°C measured for the Control C.

TABLE 3							
Property	TM*	SAMPLE					
		Control C	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Tensile Strength (MPa)	D638	16	23	17	19	21	21
Elongation (%)	D638	41	22	21	29	28	23
Tear Strength (N/mm)	D624	58	54	50	54	55	54
Max Temp (°C)	-	282	286	272	262	271	255

\* ASTM Test Method

5

Maximum temperatures for the other samples range between 255°C and 272°C. Comparing formulation Sample 2 (which includes regrind) with a similar formulation based on virgin HDPE (Sample 3), it is observed that the properties of both Sample 2 and Sample 3 formulations are very similar to each other. The maximum temperature is lower for Sample 2 and Sample 3 than is the temperature for the Control C. This result may be attributable to the enhanced fire retardancy imparted to the corresponding compositions by the combination of antimony oxide and the titanium dioxide. Similar results are obtained for formulation Sample 5 (using regrind) as for the Sample 4 control formulation (using virgin HDPE).

10

15

The intumescent composite material has certain physical properties, as presented above, which impart desirable characteristics to moldings fabricated from the intumescent composite material. These physical properties include tensile strengths, elongation ratios, and tear strength values substantially equivalent to moldings comprising virgin HDPE. These properties lend sufficient strength and durability to the moldings so as to render the moldings useful in aircraft, architectural, automotive, and marine applications, as well as other applications that will become obvious to those skilled in various arts. In addition to molding, the material may be extruded or vacuum formed to make parts that can be readily assembled into a final product.

Processability enhancement is evaluated by measuring the torque generated during melt mixing and rod extrusion of the mixed formulations. As seen in Table 2, the new formulations Samples 1, 2 and 5 exhibit appreciably lower torque as compared to the control formulation, which does not contain regrind, silicone powder, or titanium dioxide. Hence, the use of plastic fuel tank regrind as part of the formulation of intumescent composite material 20 assists in improving the melt processing of intumescent composite material 20.

Upon exposure to the extreme temperatures generated by a fire, the material swells to form a ceramic-like sponge, which is effective as a heat and fire barrier. Upon burning, the intumescent composite materials will not melt, but instead will form a relatively strong foam barrier of char and inorganic ceramic-like material. The intumescent composite material has fire retardant properties similar to those of intumescent composite materials fabricated from virgin HDPE; however, since regrind from plastic fuel tanks can be obtained at much lower costs than virgin HDPE, the overall cost of the inventive intumescent composite material will be lowered. Moreover, because the material provides a product for the recycling of plastic fuel tanks, it projects an environmentally friendly image on the plastic fuel tank industry.

DP-302280

The following examples, which are meant to be exemplary, not limiting, illustrate methods of manufacturing some of the various embodiments of the intumescent composite material using various apparatus.

## EXAMPLES

## EXAMPLE 1

On a laboratory scale, the mixing of the samples (e.g., Samples 1  
5 - 5) was achieved using a two-roll mill, as stated above. The mill was heated to  
65°C and charged with regrind or virgin HDPE, accordingly, and CPE, as well as  
with chlorowax, DSTDP, phenol, antimony oxide, pentaerythritol, and the  
phosphate-based foaming agent, and shear mixed for about 5 minutes. Good  
mixing was visually observed, and the material was banded on one of the rolls of  
10 the mill. Due to the shearing of the resins as they were mixed, the temperature of  
the material at this point approached 150°C. Hydrated magnesia and hydrated  
alumina were then added and mixed well with the material. The filler was then  
added and mixed well into the material for about 3 minutes. The total mixing  
time was about 15 minutes conducted at about 150°C. To remove the sample  
15 from the mill, a knife cut was introduced across the whole width of the sample as  
it lied on the hot roll. One cut edge was grabbed and the sample was peeled off  
the mill roll as the latter rotated at a controlled low speed.

## EXAMPLE 2

20 In another embodiment (not shown in Table 2), regrind,  
chlorowax, DSTDP, phenol, antimony oxide, pentaerythritol, and the phosphate-  
based foaming agent may be added to a mill preheated to 65°C. Silicone rubber  
may be added in place of the CPE. Upon mixing, the hydrated magnesia and  
hydrated alumina should be added as in Example 1 above. The material is then  
25 mixed, the filler added, and the mixture is mixed again. The characteristics and  
properties of the finished product will be similar to those of Example 1. It is  
expected that the resulting intumescent material will have good fire shielding  
behavior, improved processability, lower smoke generation during fire, but poor  
mechanical properties.

DP-302280

## EXAMPLE 3

- The samples were also prepared using a Banbury internal mixer. The cavity of the mixer was heated to 93°C and then charged with regrind or virgin HDPE, accordingly, and CPE, DSDTP, antimony oxide, titanium dioxide, pentaerythritol, and chlorowax. The materials were then mixed for 3 minutes at 120 rpm. The temperature, due to the shearing of the resin, approached 120°C, thus allowing the regrind to melt. The temperature was not, however, allowed to exceed 140°C to prevent degradation of the product. The foaming agent, hydrated magnesia, hydrated alumina, and the filler were then added to the mixer and mixing was continued for 3 minutes or until the temperature reached 160°C, whichever occurred first. The mix was then removed from the bowl and transferred to a two-roll mill to further mix and sheet out the composition. This demonstrates that an internal mixer can be used to make the product.
- Commercially, large batches (about 2000 lb.) can be made using the Banbury mixer. Mixing on the mill is not economical for making large batches. However, the mill is still used to dump batches of Banbury mixed batches for sheeting the product.

## 20 EXAMPLE 4

- On a plant scale, Sample 1 was prepared by mixing the material using a 3.5 inch diameter Buss Kneader having an L/D ration of 11 to 1. All ingredients except the glass fiber were dry blended and introduced into the extruder at the hopper. The glass fiber was introduced separately through a port placed downstream on the extruder barrel in order to prevent excessive pulverization. The temperature at different zones of the kneading section ranged from 138°C at the throat to 158°C at the die, through which it was extruded. At the die face, ten rods 4 mm in diameter were extruded and pelletized. Natural air cooling at the die surface was sufficient to cool the hot compound and allow

DP-302280

pelletization. No additional cooling was needed. The properties of the finished product are very similar to properties of samples prepared in the laboratory, shown in Table 3.

For samples incorporating the cross-linking agents (dicumyl  
5 peroxide and TMPT), the preparation was similar to the preparations of those in the above Examples, with the cross-linking agents being added during the last minute of mixing when the samples are prepared using the mill or the Banbury procedures. Sometimes curatives (cross-linking agents) are added on the mill during sheeting. For extruder mixing the curatives are blended in with the other  
10 ingredients and introduced at the hopper.

The present intumescent material is an environmentally friendly manner of producing plastic articles. By recycling these materials, disposal and landfill costs and issues are eliminated, and manufacturing costs are decreased.

While preferred embodiments have been shown and described,  
15 various modifications and substitutions may be made thereto without departing from the spirit and scope of the invention, including the use of the geometries taught herein in other conventional sensors. Accordingly, it is to be understood that the apparatus and method have been described by way of illustration only, and such illustrations and embodiments as have been disclosed herein are not to  
20 be construed as limiting to the claims.

DP-302280

## CLAIMS:

What is claimed is:

1. A moldable intumescent composite material, comprising,  
based upon the total weight of the composite material:  
about 2 wt% to about 60 wt% regrind;  
5 up to about 60 wt% of a first material selected from the group  
consisting of chlorinated polyethylene, silicone rubber, and combinations  
thereof;  
up to about 40 wt% foaming agent; and  
up to about 28 wt% char forming compound.
2. The moldable intumescent composite material of Claim 1,  
wherein the regrind is HDPE obtained from plastic fuel tanks.
3. The moldable intumescent composite material of Claim 1,  
further comprising about 5 wt% to about 45 wt% of the regrind, about 1 wt% to  
about 60 wt% of the first material, about 2 wt% to about 37 wt% of the foaming  
agent, and about 0.5 to about 21 wt% of the char forming compound.
4. The moldable intumescent composite material of Claim 3,  
further comprising about 15 wt% to about 25 wt% of the regrind, about 2 wt% to  
about 38 wt% of the first material, about 7 wt% to about 33 wt% of the foaming  
agent, and about 1 to about 15 wt% of the char forming compound.



DP-302280

5. The moldable intumescent composite material of Claim 1, further comprising about 5 wt% to about 60 wt% of the regrind, up to about 15 wt% plasticizer, up to about 14 wt% smoke reducing agent, up to about 2 wt% cross-linking agent, about 0.1 to about 10 wt% antioxidant, and up to about 20 wt% filler.

6. The moldable intumescent composite material of Claim 5, further comprising about 3 to about 12 wt% plasticizer, about 0.5 to about 10 wt% smoke reducing agent, up to about 1 wt% cross-linking agent, about 0.2 to about 2 wt% antioxidant, and about 2 to about 18 wt% filler.

7. The moldable intumescent composite material of Claim 6, further comprising about 5 to about 10 wt% plasticizer, about 1 to about 8 wt% smoke reducing agent, up to about 0.1 wt% cross-linking agent, and about 0.4 to about 1.2 wt% antioxidant, and about 5 to about 10 wt% filler.

8. The moldable intumescent composite material of Claim 5, wherein the smoke-reducing compound is selected from the group consisting of antimony oxide, titanium dioxide, silicone rubber, and combinations comprising at least one of the foregoing.

9. The moldable intumescent composite material of Claim 5, wherein the plasticizer is a chlorinated wax compound.

10. The moldable intumescent composite material of Claim 5, wherein the filler material is selected from the group consisting of glass, mica, carbon, titanium dioxide, clay, calcium carbonate, and combinations comprising at least one of the foregoing filler materials.

DP-302280

11. The moldable intumescent composite material of Claim 1, wherein the foaming agent is selected from the group consisting of ammonium dihydrogen-ortho-phosphate, ammonium polyphosphate, hydrated metal oxides, metal hydroxides, and combinations comprising at least one of the foregoing.

12. The moldable intumescent composite material of Claim 11, wherein the hydrated metal oxide is selected from the group consisting of hydrated alumina, hydrated magnesia, and combinations comprising at least one of the foregoing hydrated metal oxides.

13. The moldable intumescent composite material of Claim 1, wherein the char formation material is selected from the group consisting of carbohydrates, polyhydric alcohols, antimony oxide, and combinations comprising at least one of the foregoing materials.

14. The moldable intumescent composite material of Claim 13, wherein the polyhydric alcohol is a pentaerythritol.

15. The moldable intumescent composite material of Claim 1, wherein the regrind further comprises about 2 wt% or greater contaminants.

16. The moldable intumescent composite material of Claim 15, wherein the regrind further comprises about 5 wt% to about 40 wt% of the contaminants.

17. The moldable intumescent composite material of Claim 16, wherein the regrind further comprises about 5 wt% to about 15 wt% of the contaminants.

DP-302280

18. The moldable intumescent composite material of Claim 15, wherein the contaminants are selected from the group consisting of ethylene vinyl alcohol copolymer, adhesive, fillers, and combinations comprising at least one of the foregoing contaminants.
19. The moldable intumescent composite material of Claim 15, wherein the adhesive is maleated polyethylene.
20. The moldable intumescent composite material of Claim 1, wherein the regrind comprises HDPE which was processed at temperatures exceeding about 150°C.
21. The moldable intumescent composite material of Claim 20, wherein the regrind comprises HDPE which was processed at temperatures of about 150°C to about 190°C.
22. An article formed from the moldable intumescent composite material of Claim 1.
23. A moldable intumescent composite material, comprising the reaction product of, based upon the total weight of the composite material:
- about 2 wt% to about 60 wt% regrind;
  - up to about 60 wt% of a first material selected from the group
  - 5 consisting of chlorinated polyethylene, silicone rubber, and combinations thereof;
  - up to about 40 wt% foaming agent; and
  - up to about 28 wt% char forming compound.

DP-302280

24. The moldable intumescent composite material of Claim 23, wherein the regrind further comprises about 2 wt% or greater contaminants.

25. The moldable intumescent composite material of Claim 24, wherein the regrind further comprising about 5 wt% to about 40 wt% of the contaminants

26. The moldable intumescent composite material of Claim 25, wherein the regrind further comprising about 5 wt% to about 15 wt% of the contaminants.

27. The moldable intumescent composite material of Claim 24, wherein the contaminants are selected from the group consisting of ethylene vinyl alcohol copolymer, adhesive, fillers, and combinations comprising at least one of the foregoing contaminants.

28. The moldable intumescent composite material of Claim 27, wherein the adhesive is maleated polyethylene.

29. The moldable intumescent composite material of Claim 23, wherein the regrind comprises HDPE which was processed at temperatures exceeding about 150°C.

30. The moldable intumescent composite material of Claim 23, wherein the regrind comprises HDPE which was processed at temperatures of about 150°C to about 190°C.

DP-302280

INTUMESCENT MATERIAL USING HIGH DENSITY POLYETHYLENE  
REGRIND

ABSTRACT OF THE INVENTION

A moldable intumescent composite material comprises ground recycled plastic, chlorinated polyethylene, a plasticizer, a foaming agent, a char formation compound, an antioxidant, hydrated oxides, smoke-reducing compounds, and filler material. The ground recycled plastic is typically HDPE  
5 obtained from plastic fuel tanks. The smoke-reducing compounds are antimony oxide, titanium dioxide, or a combination thereof. Silicone rubber may also be utilized as a smoke-reducing compound. A cross-linking agent may further be incorporated into the material to improve the rigidity of the material when it is exposed to fire. The invention also includes a method of improving the  
10 processability of the intumescent composite material during formulation. The method comprises adding titanium dioxide and silicone rubber to the material and molding the material to a desired shape.

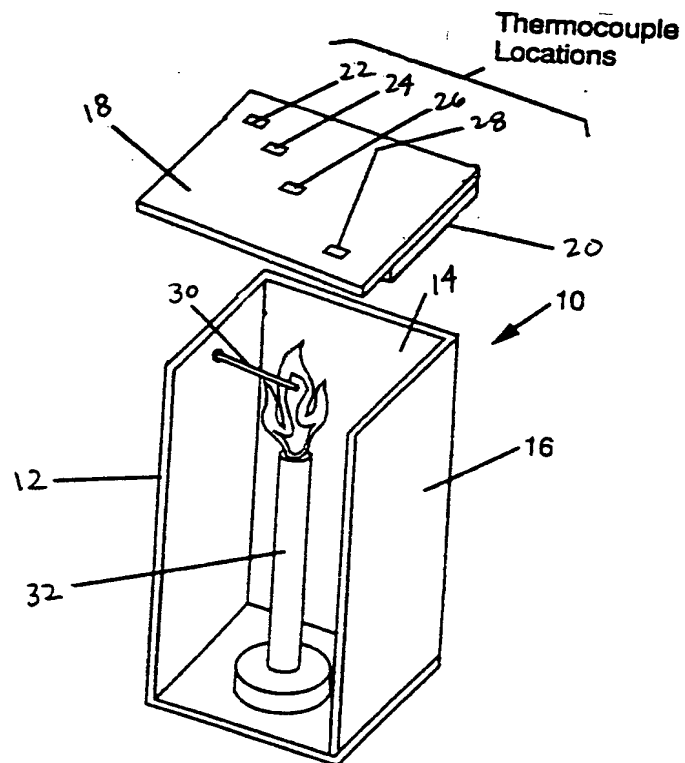


FIG. 1

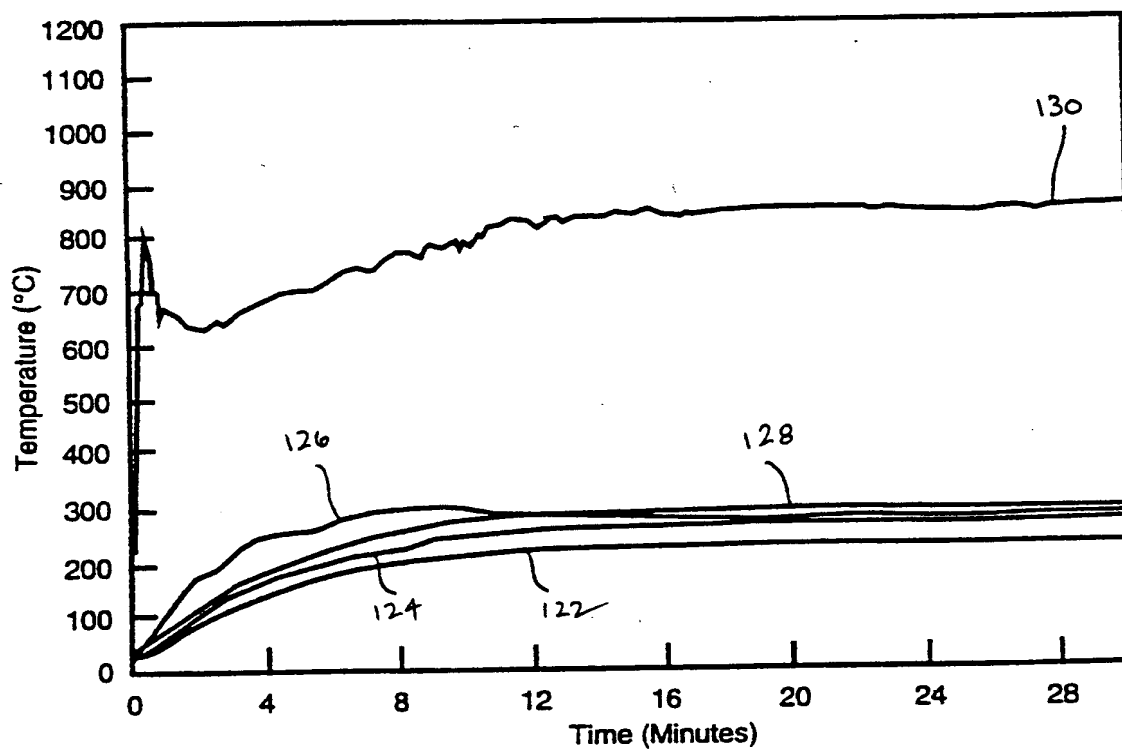


FIG. 2

Parts List:

Wall	12, 14, 16
Roof	18
Material	20
Locations	22, 24, 26, 28
Thermocouple	30
Bunsen burner	32